Magnetic Properties of RIrSi (R = Tb, Dy, and Ho) Compounds

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(Received December 12, 2012)

The magnetic data for the polycrystalline samples of RIrSi (R = Tb, Dy and Ho) compounds are reported. These compounds are antiferromagnets with the Néel temperatures equal to 32 K (R = Tb), 7.0 K (R = Dy) and 4.8 K (R = Ho), respectively. The external magnetic field induces the one step for TbIrSi and two step for DyIrSi and HoIrSi metamagnetic phase transitions. The values of the critical field as Néel temperatures decrease with increase of the number of $4f$ electrons. The magnetic phase diagrams (H, T) are determined.

DOI: 10.12693/APhysPolA.123.773

PACS: 75.30.Kz, 75.40.Gb, 75.47.Np, 75.50.Ee

1. Introduction

As a continuation of the investigations of the magnetic properties of the ternary rare earth intermetallics this work reports the results for RIrSi (R = Tb, Dy, and Ho) compounds. These compounds crystallize in the orthorhombic crystal structure of the TiNiSi-type (space group *Pnma*) [1–3]. Magnetic measurements at low magnetic field indicate that these compounds are antiferromagnets with the Néel temperature equal to 32 K (R = Tb), 7 K (R = Dy) and 4.8 K (R = Ho). The neutron diffraction data for Tb- and Ho-compounds confirm the sine modulated and collinear antiferromagnetic structure, respectively. For TbIrSi this order is stable in the temperature range 1.5 K–T_{N}. The temperature dependence of the diffraction data for HoIrSi indicate below T_{N} at $T = 4.5$ K a coexistence of the collinear magnetic structure and the sine modulated one [3].

In purpose to explanation the magnetic properties of these compounds at low temperatures the new dc and ac magnetic measurements are performed.

2. Experimental

RIrSi (R = Tb, Dy and Ho) polycrystalline samples were synthesized by arc melting stoichiometric amounts of high-purity elements (R of 3N purity, Ir of 4N purity and Si of 5N purity).

The X-ray powder diffraction patterns were recorded at room temperature using a PANalytical X'Pert PRO MPD diffractometer (Cu K_α radiation). The data were analyzed by the Rietveld-type line profile refinement method using the program Fullprof [4].

The magnetic measurements were carried out using a vibrating sample magnetometer (VSM) option of the Quantum Design PPMS platform. Three types of magnetic measurements were performed: cooling at low temperatures at zero magnetic field (ZFC) and at field (FC) equal to $H = 50$ Oe (to determine the phase transition temperatures), then scanning room 1.9 K up to 300 K in a magnetic field of 1 kOe (to determine the values of the effective magnetic moment μ_{eff} and the paramagnetic Curie temperatures θ_{p}), and finally measuring the magnetization curves up to 90 kOe between the Néel temperatures 1.9 K (to determine the values of the magnetic moment in the ordered state and magnetic phase diagrams).

For all compounds the temperature dependence of the ac magnetic susceptibility $\chi_{\mathrm{ac}} = \chi' - \mathrm{i}\chi''$, where χ' is the real and χ'' the imaginary component, was measured at several chosen frequencies between 10 Hz and 10 kHz in the temperature range 2–46 K.

3. Results

The analysis of the X-ray data indicates that all compounds have the orthorhombic crystal structure of TiNiSi-type described by the space group *Pnma*.

The results of the dc magnetic measurements for all three compounds are shown in Fig. 1. The reciprocal magnetic susceptibility of all investigated compounds obeys the Curie–Weiss law. The paramagnetic Curie temperature change from negative value (−10.3 K) for TbIrSi by near to zero (0.2 K) for DyIrSi to positive (3.8 K) for HoIrSi. The values of the effective magnetic moments are close to R^{3+} ion values (see Table).

At low temperatures (see upper insets) for TbIrSi three anomalies at 2.7, 32, and 43.5 K and one at 7.0 K for DyIrSi and 4.8 K for HoIrSi are observed. For all compounds the difference between FC and ZFC curves are observed. These divergences are observed above the maximum corresponding to the Néel temperature at 43.5 K (R = Tb), 15 K (R = Dy) and 12 K (R = Ho).

Temperature dependence of the real χ' and imaginary χ'' component of the ac magnetic susceptibilities are

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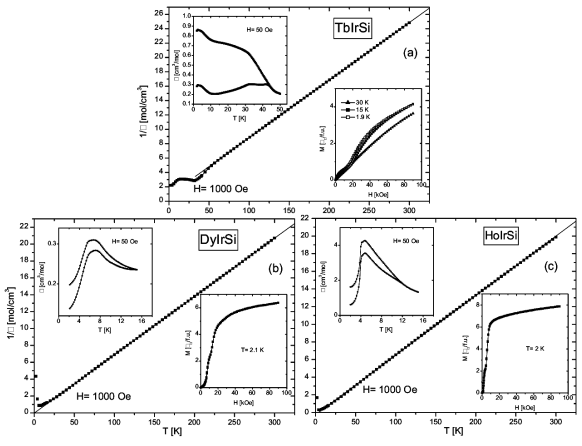

Fig. 1. Temperature dependence of the reciprocal magnetic susceptibility measured at 1 kOe magnetic field for (a) TbIrSi, (b) DyIrSi, and (c) HoIrSi. Insets show: the upper ones — temperature dependence of the magnetic susceptibilities at low temperatures: ZFC (lower curve) and FC (upper curve) and lower ones — magnetization curves up to 90 kOe at 1.9, 15, and 35 K for TbIrSi and 1.9 K for DyIrSi and HoIrSi.

shown in Fig. 2. For TbIrSi (Fig. 2a) temperature dependence of the χ' give the two maxima first at 31.5 K and second at low temperatures which change from 2.7 K for $f = 100$ Hz to 3.4 K for 10 kHz. The anomaly at 43 K was not observed. In the temperature dependence of the χ'' the large increase of values is observed below 4 K. The values of $\chi''(T)$ slowly increase with increase of the frequency. For all frequencies the small maximum near 44 K is observed. With increase of the frequency at 2.7 and 5.0 kHz the small maximum at 31.5 K is detected. The maximum at 2.7 K is probably connected with the purity phase Tb–O.

For DyIrSi in $\chi'(T)$ the broad maximum at 7 K is observed (see Fig. 2b). The values of $\chi''(T)$ increase with increase of the frequency. In $\chi''(T)$ the small broad maximum for the frequency above 500 Hz at $T = 7$ K are observed. For the frequency equal to 10 kHz the additional maximum at 4 K is observed. For HoIrSi in $\chi'(T)$ the maximum at 4.8 K is observed, whereas in $\chi''(T)$ the two maxima at 4.8 and 6.8 K are detected (see Fig. 2c). The positions of these maxima do not change in the function of the frequency.

TABLE

Magnetic data for RIrSi (R = Tb, Dy and Ho) compounds: the Néel temperature (T_N), the transition temperature (T_t), paramagnetic Curie temperature (θ_p), experimental (exp.) and theoretical (theor.) values of the effective magnetic moment (μ_{eff}) and in the ordered state (μ) at low temperatures and $H = 90$ kOe and the magnetic critical field (H_{cr}). Methods (M): static (dc) and dynamic (ac) susceptibilities.

R	M	T_N [K]	T_t [K]	θ_p [K]	μ_{eff} [μ_B]		μ [μ_B]		H_{cr} [kOe]	Ref.
					exp.	theor.	exp.	theor.		
Tb	dc	43.5, 32	2.7, 9.5	−10.3	10.0	9.72	4.15	9.0	25	this work
	ac	31.5								this work
	dc	32					8.96(16)*			[3]
Dy	dc	7.0		0.2	10.77	10.65	6.4	10.0	15, 8	this work
	ac	7.0								this work
	dc	7.0								[3]
Ho	dc	4.8		3.8	10.9	10.61	7.9	10.0	5.5, 1.6	this work
	ac	4.8								this work
	dc	4.6					9.9(1)*			[3]

* The neutron diffraction data at $T = 1.5$ K [3].

The magnetization curves at 1.9 K (see lower insets) indicate the metamagnetic phase transitions with the one critical field equal to 24 kOe for TbIrSi (inset in Fig. 1a) and two critical magnetic fields at 8.0 and 15 kOe for DyIrSi (inset in Fig. 1b) and 1.6 and 5.2 kOe for HoIrSi (inset in Fig. 1c). For TbIrSi on magnetization curve at 15 K the metamagnetic phase transition is observed whereas at 35 K magnetization is linear function of the magnetic field. The values of the critical field are determined from the relation dM/dH vs. H. For TbIrSi one maximum is observed. The determined values of the critical field are equal to 25 kOe and do not change with increase of the temperature. For HoIrSi dM/dH dependence up to $T = 3.9$ K (see Fig. 3) indicates the two maxima corresponding to the critical field H_{c1} and H_{c2}, while at $T = 4.5$ K only anomaly at H_{c2} is observed. The determined magnetic phase diagrams for DyIrSi and HoIrSi compounds are listed in Fig. 4. For DyIrSi and HoIrSi three magnetic phases are observed. Below H_{c1} according to the neutron diffraction data reported in Ref. [3] for HoIrSi the collinear antiferromagnetic structure is observed, where above H_{c2} probably ferromagnetic order

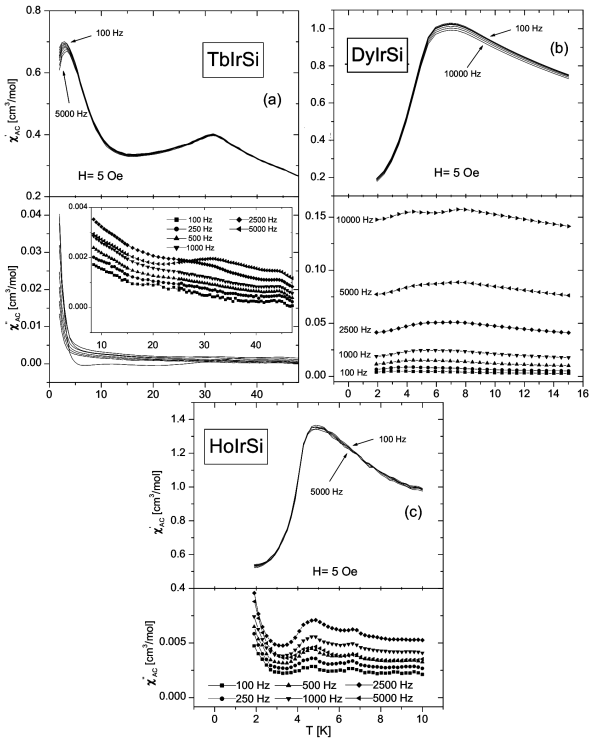

Fig. 2. Temperature dependence of the χ' and χ'' component of the ac magnetic susceptibility for (a) TbIrSi, (b) DyIrSi and (c) HoIrSi.

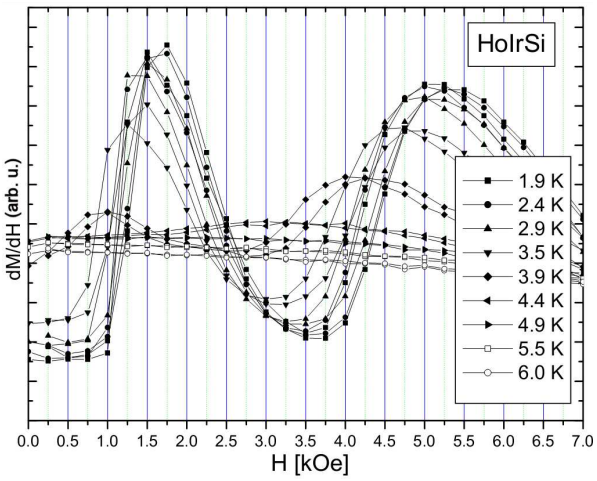

Fig. 3. The magnetic field dependence of the magnetic susceptibility $\frac{\mathrm{d}M}{\mathrm{d}H}$ for HoIrSi measured at the different temperatures.

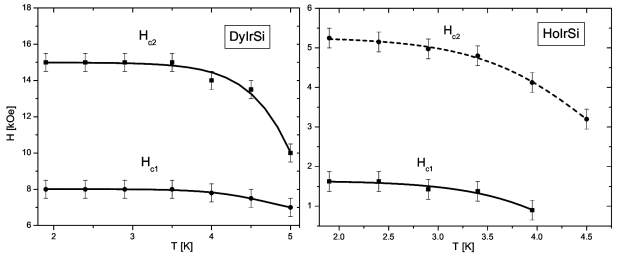

Fig. 4. Magnetic phase diagrams (H, T) for (a) DyIrSi and (b) HoIrSi.

or complex order with the ferromagnetic component is stable. Phases between H_{c1} and H_{c2} characterizing the small values of magnetization are equal to 2.1 μ_B/f.u. for DyIrSi and 2.2 μ_B/f.u. for HoIrSi. The magnetization not saturated in the field $H = 90$ kOe and determine values of the magnetic moments per f.u. are smaller than free R^{3+} ion and determine from the neutron diffraction experiment [3] values (see Table).

4. Discussion

The data presented in this work indicate that all investigated compounds are antiferromagnets at low temperatures. The presented results are generally in good agreement with the previous data [3] concerning the values of the Néel temperature. The determined values of the effective magnetic moments equal to the free R^{3+} ion values indicate that the magnetic moments are localized on the rare-earth atoms. This result is in good agreement with the neutron diffraction data [3]. The new data are observed — the large difference between the FC and ZFC curves also above and below the Néel temperature which indicate the short-range order above T_N. Large difference ZFC and FC curves below T_N for TbIrSi indicate that the magnetic field strongly influence on the modulated order. For DyIrSi and HoIrSi the smaller difference between ZFC and FC curves suggest the small influence of the magnetic field on the collinear order.

Magnetization curves indicate the metamagnetic phase transitions and do not saturate at the magnetic field of 90 kOe. For DyIrSi and HoIrSi the increase of the value of the magnetization in the function of the magnetic field has a two-step character while for TbIrSi on-step magnetization processes are observed. These indicate that in DyIrSi and HoIrSi three successive magnetic phases occur. According to the neutron diffraction data for HoIrSi below H_{cr1} the collinear antiferromagnetic ordering described by the propagation vector $\boldsymbol{k} = (1/2,\ 0,\ 1/2)$ is stable [3]. The Ho moments in a–c plane form order described by the C-vector with the sequence $+\ +\ -\ -$ in crystal unit cell (see Fig. 5b in Ref. [3]) and are parallel to the short b-axis. With increase of the magnetic field between H_{cr1} and H_{cr2} the new phase with the moment equal to 2.2 μ_B/f.u. which is 2/7 of those at 90 kOe is observed. Above H_{cr2} the ferromagnetic order or order with the ferromagnetic component exists. Similar field dependence of the magnetization in DyIrSi compound is observed.

For TbIrSi with the modulated magnetic structure [3] one-step metamagnetic process is detected. This process is observed at higher magnetic field than for isostructural Dy and Ho-compounds and has different character.

Similar one-step metamagnetic process is observed in the modulated phase of HoAlGa [5].

The magnetization process is similar to those observed in large number of the rare earth intermetallics and satisfactorily interpreted in simple model involving a huge uniaxial anisotropy and exchange interactions [6, 7].

Additional anomalies observed in the temperature and frequency dependencies of the imaginary component of the ac magnetic susceptibility indicate the relaxation process in these compounds.

Acknowledgments

The research was carried out with the equipments purchased thanks to the financial support of the European Regional Development Fund in the framework of the Polish Innovation Economy Operational Program (contract no. POIG.02.01.00-12-023/08).

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